

A Discrete Solid-on-Solid Model for Nonequilibrium Growth Under Surface Diffusion Bias

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A limited mobility nonequilibrium solid-on-solid dynamical model for kinetic surface growth is introduced as a simple description for the morphological evolution of a growing interface under random vapor deposition and surface diffusion bias conditions. Large scale stochastic Monte Carlo simulations using a local coordination dependent instantaneous relaxation of the deposited atoms produce complex surface morphologies whose dynamical evolution is not consistent with any of the existing continuum dynamical surface growth equations. Critical exponents for coarsening and roughening dynamics of the morphological evolution are quantitatively calculated using large scale simulations.

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An atom moving on a free surface is known to encounter an additional potential barrier, often called a surface diffusion bias [1] (or an Ehrlich [2] - Schwoebel [3] barrier), as it approaches a step from the upper terrace – there is no such extra barrier for an atom approaching the step from the lower terrace (the surface step separates the upper and the lower terraces). Since this diffusion bias makes it preferentially more likely for an atom to attach itself to the upper terrace than the lower one, it leads to mound (or pyramid) - type structures on the surface under growth conditions as deposited atoms are probabilistically less able to come down from upper to lower terraces. This dynamical growth behavior under a surface diffusion bias is sometimes called an “instability” because a flat two dimensional surface growing under a strong surface diffusion bias is unstable toward three dimensional mound/pyramid formation. There has been a great deal of recent interest [1,4–20] in the morphological evolution of growing interfaces under nonequilibrium growth conditions in the presence of such surface diffusion bias. In this Letter we introduce, what we believe to be, the *minimal* nonequilibrium atomistic growth model for ideal molecular beam epitaxial - type random vapor deposition growth under a surface diffusion bias. Extensive stochastic simulation results presented in this paper establish the morphological evolution of a surface growing under diffusion bias conditions to be rather complex. Various critical growth exponents [21,22], which asymptotically describe the large-scale dynamical evolution of the kinetically growing surface in our minimal discrete growth model, are inconsistent with *all* the proposed continuum theories for nonequilibrium surface growth under diffusion bias conditions. Our results lead to the conclusion that a continuum description for nonequilibrium growth under a surface diffusion bias does not exist and may require a theoretical formulation which is substantially different from the ones currently existing in the literature.

In Fig. 1(a) we schematically describe our solid-on-solid (SOS) nonequilibrium growth model : (1) Atoms

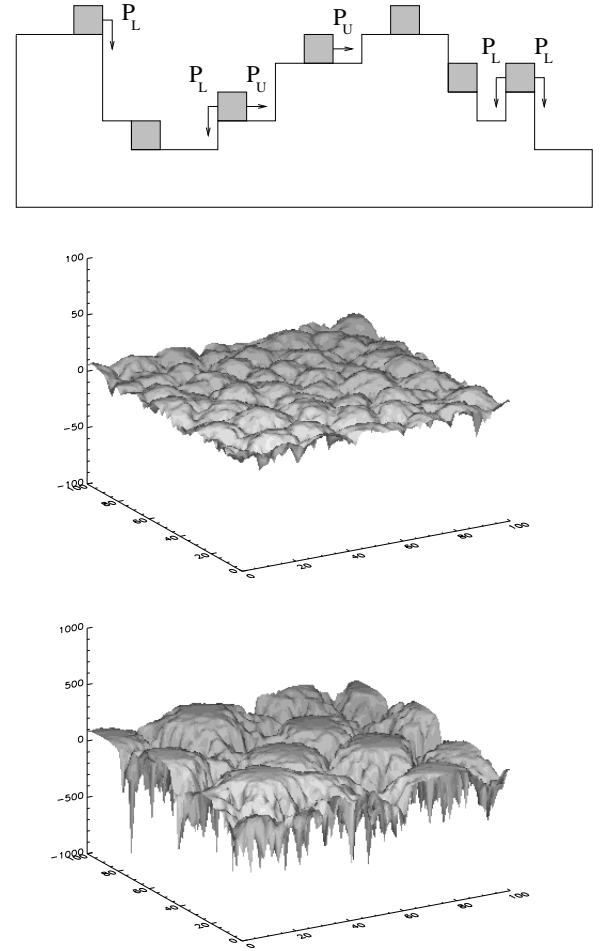


FIG. 1. (a) Schematic configuration defining growth rules in 1+1 dimensions. (b) Morphologies ($P_U = 1; P_L = 0.5$) for $L = 100 \times 100$ at 10^3 ML and (c) 10^6 ML.

are deposited randomly (with an average rate of 1 layer/unit time, which defines the unit of time in the growth problem – the length unit is the lattice spacing taken to be the same along the substrate plane and the growth direction) and sequentially on the surface starting with a flat substrate; (2) a deposited atom is incorporated instantaneously if it has at least one lateral nearest - neighbor atom (i.e. if it has a coordination of 2 or more since there must always be an atom underneath to satisfy the SOS constraint); (3) singly coordinated deposited atoms (i.e. the ones without any lateral neighbors) could instantaneously relax to a neighboring site within a diffusion length of l provided the neighboring site of incorporation has a higher coordination than the original deposition site; (4) the instantaneous relaxation process is constrained by two probabilities P_L and P_U (with $0 \leq P_L, P_U \leq 1$) where $P_{L(U)}$ is the probability for the atom to attach itself to the lower(upper) terrace after relaxation (note that a “terrace” here could be just one other atom). The surface diffusion bias is implemented in our model by simply taking $P_U > P_L$, making it more likely for atoms to attach to the upper terrace. Under the surface diffusion bias, therefore, an atom deposited at the top of a step edge feels a barrier (whose strength is controlled by $P_U - P_L$) in coming down compared with an atom at the lower terrace attaching itself to the step. Our surface diffusion bias model is well-defined for any value of the diffusion length l including the most commonly studied situation of nearest - neighbor relaxation ($l = 1$). We have carried out extensive simulations both in 1+1 and 2+1 dimensions (d) varying P_L , P_U as well as l , also including in our simulations the inverse situation (the so-called ‘negative’ bias condition) with $P_L > P_U$ so that deposited atoms preferentially come down attaching themselves to lower steps producing in the process a smooth growth morphology. Because of lack of space we do not present our negative bias results here except to note that the smooth dynamical growth morphology under our negative bias model obeys the generic Edwards - Wilkinson universality [1,21,22].

Before presenting our numerical results we point out two important features of our growth model : (1) For $P_L = P_U = 1$ our model reduces to the one introduced in ref. 23 (and studied extensively [21–27] in the literature) as a minimal model for molecular beam epitaxy in the absence of any diffusion bias; (2) we find, in complete agreement with earlier findings [23,24] in the absence of diffusion bias, that the diffusion length l is an *irrelevant* variable which does not affect any of our calculated critical growth exponents (but does affect finite size corrections – increasing l requires a concomitant increase in the system size to reduce finite size effects). We, therefore, mostly present our $l = 1$ simulation results here emphasizing that our critical exponents are independent of l provided finite size effects are appropriately accounted for. Our calculated exponents are also independent of the precise values of P_U and P_L as found in ref. 23 and 24 for the $P_U = P_L$ case.

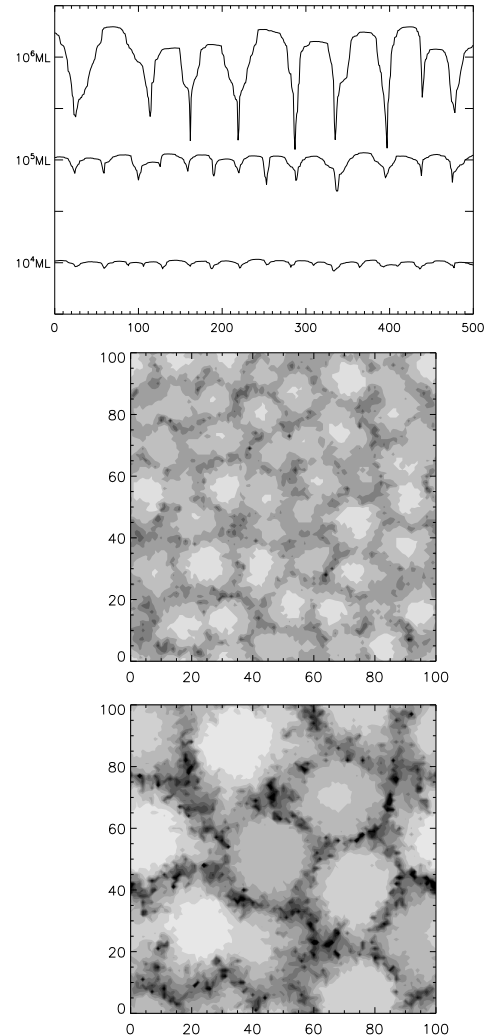


FIG. 2. (a) The mound evolution in 1+1 dimensions showing a section of 500 middle lattice sites from a substrate size of $L = 10000$ at 10^4 ML, 10^5 ML, and 10^6 ML ($P_U = 1$; $P_L = 0.5$). (b,c) The d=2+1 growth morphologies on a 100×100 substrate (b) 10^3 ML, and (c) 10^6 ML. The darker (lighter) shades represent lower (higher) points on the surface.

In Fig.1 we show representative d=2+1 simulated growth morphologies for our positive bias model. The diffusion bias produces mounded structures which are visually statistically scale invariant only on length scales much larger (or much smaller) than the typical mound size. In Fig.2 we show morphological evolutions for d=2+1 and 1+1 in order to visualize the coarsening/steepening dynamics of the mound structures. We mention that in producing our final results we utilize a noise reduction [28] technique which accepts only a fraction of the attempted kinetic events, and in the process produces smoother results (reducing noise effects) without in any way affecting the critical exponents. The corresponding results without noise reduction are visually more noisy with identical growth exponents.

To proceed quantitatively we now introduce the dynamic scaling ansatz which seems to describe well all our simulated results. We have studied the root mean square surface width or surface roughness (W), the average mound size (R), the average mound height (H), and the average mound slope (M) as functions of growth time. We have also studied the various moments of dynamical height-height correlation function, and these correlation function results (to be reported elsewhere) are consistent with the ones obtained from our study of $W(t)$, $R(t)$, $H(t)$, and $M(t)$. The dynamical scaling ansatz in the context of the evolving mound morphologies can be written as power laws in growth time (which is equivalent to power laws in the average film thickness) : $W(t) \sim t^\beta$; $R(t) \sim t^n$; $H(t) \sim t^\kappa$; $M(t) \sim t^\lambda$; $\xi(t) \sim t^{1/z}$, where $\xi(t)$ is the lateral correlation length (with z as the dynamical exponent) and β , n , κ , λ , z are various growth exponents which are not necessarily independent. In the steady state, when $\xi(t) \gtrsim L$ where L is the lateral substrate size, effective β vanishes as the surface roughness saturates to a value $W_s(L) \equiv W(L, t \rightarrow \infty) \sim L^\alpha$, where $\alpha = \beta z$ is the roughness exponent. We find in all our simulations $n \approx z^{-1}$, and thus the coarsening exponent n , which describes how the individual mound sizes increase in time, is the same as the inverse dynamical exponent in our model. We also find $\beta = \kappa$ in all our results, which is understandable in a mound-dominated morphology. In addition, all our results satisfy the expected exponent identity $\beta = \kappa = n + \lambda$ because the mound slope $M \sim H/R$. The evolving growth morphology is thus completely defined by two independent critical exponents β (the growth exponent) and n (the coarsening exponent), which is similar to the standard (i.e. without any diffusion bias) dynamic scaling situation where β and z ($= n^{-1}$ in the presence of diffusion bias) completely define the scaling properties. We note also that our *negative* bias results (not shown here for lack of space) are completely consistent with the expected [1,22] Edwards - Wilkinson universality class [1,21,22] with our numerical findings being $\beta = 0.25$ and $z = 2$ in $d=1+1$, and $\beta = 0$ (i.e. $W \sim \ln t$) and $z = 2$ in $d=2+1$. This is expected because our negative bias model explicitly introduces an inclination dependent downhill surface current [29] in the problem.

In Fig.3 we show our representative scaling results for nonequilibrium growth under surface diffusion bias conditions. It is clear that we consistently find $\beta \simeq 0.5$ in both $d=1+1$ and $2+1$ for growth under a surface diffusion bias. This $\beta \simeq 0.5$ is, however, very different from the usual Poisson growth under pure random deposition with no relaxation where there are no lateral growth correlations. Our calculated asymptotic coarsening exponent n in both $d=1+1$ and $2+1$ is essentially zero (< 0.1) at long times. In all our results we find the effective coarsening exponent showing a crossover from $n \approx 0.2$ at early times to a rather small value (< 0.1) at long times – we believe the asymptotic n to be zero in our model.

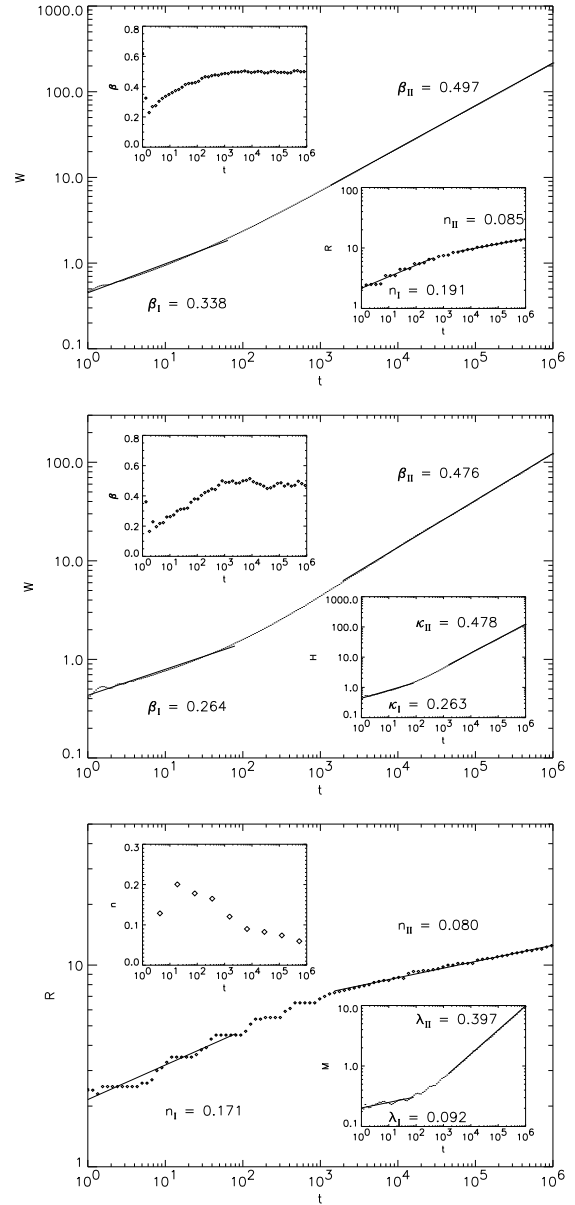


FIG. 3. (a) The surface roughness W in 1+1 dimensions as a function of deposition time t in the $L = 10000$ system. Left inset: the growth exponent β calculated from the local derivative of $\log_{10} W$ with respect to $\log_{10} t$. Right inset: Average mound size as a function of deposition time. (b) The surface roughness in the 100×100 system. Left inset: the local growth exponent β . Right inset: Average mound height vs time. (c) The average mound size in the 100×100 system. Left inset: the local coarsening exponent n calculated from the local derivative of $\log_{10} R$ with respect to $\log_{10} t$. Right inset: the average mound slope vs time.

In comparing with the existing continuum growth equation results (of which there are quite a few) we find that none can quantitatively explain all our findings. Golubovic [18] predicts $\beta = 0.5$, which is consistent with our simulations, but his finding of $n = \lambda = 1/4$ in both $d=1+1$ and $2+1$ is inconsistent with our results ($n < 0.1$, $\lambda \simeq 0.4-0.5$) except in the limited transient regime. The analytic results of Rost and Krug [19] also cannot explain our results, because they predict, in agreement with Golubovic, that if $\beta = 1/2$, then $n = \lambda = 1/4$, which is in disagreement with our findings. We also find our β to be essentially 0.5 independent of the actual value of $P_U - P_L$, which disagrees with ref. 19. Our model obviously has no slope selection ($\lambda \equiv 0$), and therefore theories predicting slope selections do not apply.

Before concluding we point out some reasons for why we believe our nonequilibrium limited mobility growth model to be a good zeroth order description for kinetic growth under a surface diffusion bias. The main reason for this is the success of the corresponding minimal growth model, introduced in ref. 23, in providing a good zeroth order description of molecular beam epitaxial growth in the absence of any surface diffusion bias. The $d=2+1$ critical exponents [27] in the unbiased model [23] are $\beta = 0.25 - 0.2$ and $\alpha \simeq 0.6 - 0.7$, which are in quantitative agreement with a number of experimental measurements [21,22] where surface diffusion bias is thought to be dynamically unimportant. An equally significant theoretical reason is that the corresponding unbiased growth model [23–27] is the *only existing* nonequilibrium growth model which is known [22,27] *not* to have the linear Edwards - Wilkinson ‘ $\nabla^2 h$ ’ term [1] in its coarse-grained long wavelength continuum description (an equivalent statement [29] is that this is the only limited mobility model which has a vanishing surface current on a tilted substrate) – in fact, our negative bias model introduces precisely this ‘ $\nabla^2 h$ ’ term by producing a downhill current on a tilted substrate. (The other limited mobility nonequilibrium growth models [30,31] introduced in the literature are known to belong to the Edwards - Wilkinson universality class, and are therefore unsuitable for diffusion bias studies [32].) Therefore, the minimally biased version of this model which we study in this paper should be the appropriate zeroth order description for nonequilibrium growth under surface diffusion bias conditions. Since an approximate continuum description for the unbiased model [23] has recently been developed [26], one could use that as the starting point to construct a continuum growth model for the biased growth situation. Such a continuum description is, however, extremely complex [26] as it requires the existence of an infinite number of terms in the growth equation. It remains unclear that a meaningful continuum description for nonequilibrium growth under a surface diffusion bias is indeed possible even for our simple limited mobility growth model.

We conclude by mentioning that any comparison to experiments, as has been done in several recent theoretical

publications on this topic, is premature at this stage because the real Ehrlich - Schwoebel barrier in experimental systems [33] is expected [34] to be quite complicated, and simple growth models used by us and others [4–20] most likely do not apply. What we have established in this paper is that even a very simple limited mobility nonequilibrium growth model leads to extremely complex dynamical interface morphologies under surface diffusion bias conditions. The fact that even a deceptively simple limited mobility growth model such as the one studied in this paper seems to defy a theoretical continuum description is a strong indication that our understanding of nonequilibrium growth under a surface diffusion bias is far from complete.

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